

A Model for the Determination of the Scavenging Rates of Submicron Aerosols by Snow Crystals*

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ABSTRACT

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A theoretical model for the determination of the rate at which submicron aerosol particles are scavenged by snow crystals is presented. This scheme incorporates Brownian diffusion, thermophoresis, diffusiophoresis, and electrostatic attraction. Scavenging rates are derived as a function of aerosol size, snow crystal capacitance, atmospheric temperature, and relative humidity. The model applies to any snow crystal type that can be described by its capacitance and a generalized gamma distribution where the coefficients are dependent on the crystal type. The crystal surface properties are determined by applying the Gauss theorem over an arbitrary surface where a linearized capacitance is applied. Relative humidity is the most sensitive of the terms being modeled in this particle size range. A 5% change in relative humidity results in a change in the scavenging rate of up to an order of magnitude, while a 50% change in capacitance changes the scavenging rate by a factor to two. Empirical models do not include relative humidity effects. Observed data are limited but lend partial verification to this model.

RESUME

On présente un modèle théorique de détermination du taux de lessivage des particules sous-microniques par les cristaux de neige. Ce modèle prend en compte la diffusion brownienne, la thermophorèse, la diffusiophorèse, et l'attraction électrostatique. Les taux de lessivage sont calculés en fonction de la dimension de l'aérosol, de la capacitance du cristal de neige, de la température de l'air et de l'humidité relative. Le modèle s'applique à tout type de cristal de neige pouvant être décrit par sa capacitance, et par une distribution gamma généralisée dans laquelle les coefficients dépendent du type de cristal. Les propriétés de surface du cristal sont déterminées en appliquant le théorème de Gauss à une surface arbitraire où est appliquée une capacitance linéarisée. L'humidité relative est le plus sensible des paramètres testés dans cette gamme de dimensions. Un changement de 5% de l'humidité relative provoque une variation d'un ordre de grandeur dans le taux de lessivage, tandis qu'un changement de 50% de la capacitance fait varier le taux de lessivage

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d'un facteur deux. Les modèles empiriques ne prennent pas en compte les effets de l'humidité relative. Les données expérimentales sont rares, mais elles tendent partiellement à vérifier le modèle.

INTRODUCTION

Early snow scavenging field studies (Englemann and Perkins, 1966; Carnuth, 1967) indicated that snow water has concentrations of aerosol particles up to twice that of rain water at equivalent precipitation rates. Although this large scavenging difference has led to studies on snow scavenging rates (Knutson and Stockman, 1974), geometric complexities of snow crystals and lack of data on relevant parameters have severely limited theoretical studies of the rates at which aerosols are scavenged by precipitating snow crystals.

Field experiments with natural snow by Sood and Jackson (1969, 1970) and by Knutson et al. (1976) enabled Knutson and Stockham (1974) to formulate an empirical relation for the scavenging rates of submicron aerosols by snow crystals:

$$A = AP^{\alpha_1} \exp[G(T, D_p)] \quad (1a)$$

where P is the precipitation rate (mm h^{-1}), D_p (μm) is the particle diameter and T is temperature (F). The constant A (0.21 to 0.41) and α_1 (0.19 to 0.57) are empirical fits dependent on snow crystal geometry. The function $G(T, D_p)$ is defined for $D_p = 0.5 \mu\text{m}$ as

$$G(T, D_p) = 3.17 - 0.017T + 3.41 \log(D_p) - 0.973(\log D_p)^2 - 7.20(\log D_p)^3 \quad (1b)$$

This model parametrizes scavenging rates as a function of snow crystal type, particle size, and atmospheric temperature, but relative humidity and charge are omitted.

Lacaux and Warburton (1982) studied scavenging rates by introducing silver iodide ($D_p = 0.03 \mu\text{m}$) into clouds. By measuring the mass balance of silver iodide and the amount brought down to the surface in precipitation, they were able to deduce a scavenging rate of the form

$$A = -\ln(1 - E)/t, \quad (2)$$

where E is the measured collection efficiency and t is the precipitation period.

A theoretical scavenging rate has been provided by Slinn (1984). However, several assumptions and simplifications were made in arriving at a final formula. Slinn presented a snow scavenging rate dependent on the precipitation rate (P , mm h^{-1}), the equivalent snow crystal diameter (D_c), and the collection efficiency (E), with the following result:

$$A = \gamma_1 RE/D_c, \quad (3)$$

where γ_1 is a constant that is fitted to the few data points of Englemann and Perkins (1966) for the case $D_p = 0.5 \mu\text{m}$. Slinn (1977) suggested a possible form for E but did not compute its value; E was set to unity. Slinn's formula does not take into account particle size, temperature, or relative humidity.

Scavenging rates for the above formulations are between $3 \cdot 10^{-6}$ and $2 \cdot 10^{-5}$ in eq. 1 with $D_p = 0.5 \mu\text{m}$, about $1.2 \cdot 10^{-4}$ in eq. 2 for $D_p = 0.03 \mu\text{m}$, and between 10^{-6} and $2 \cdot 10^{-5}$ in eq. 3 for $D_p = 0.5 \mu\text{m}$. The larger scavenging rate in eq. 2 is expected, as Brownian diffusion is the dominant scavenging mechanism for this $0.03\text{-}\mu\text{m}$ size range. In the $0.5\text{-}\mu\text{m}$ size range thermophoresis is the dominant scavenging mechanism. In all cases the relative humidity is assumed to be nearly 100%.

To date there have been no accurate formulations of snow scavenging rates in which atmospheric temperature, relative humidity, electrical charge, Brownian diffusion, particle size, and snow crystal size are all incorporated into the model. Previous studies of the collision efficiency by which aerosols are collected by ice crystals (Wang and Pruppacher, 1980; Martin et al., 1980; Wang, 1985; Miller, 1987; Miller and Wang, 1989) indicate that for aerosols between 0.01 and $1.0 \mu\text{m}$, relative humidity plays a major role in the removal of aerosols. Decreasing relative humidity increases the scavenging rate so that a maximum occurs for the driest air with particles approaching $1.0 \mu\text{m}$ in radius. Opposing electrostatic charges between aerosols and snow crystals enhance the scavenging efficiency, as does increasing temperature and pressure. Brownian diffusion of particles onto crystal surfaces increases with decreasing submicron particle size and increasing snow crystal surface area-to-volume ratio. The mathematical formulation of this theory extended to scavenging rates is presented in the next section.

THEORY

The mathematical form of the rate of scavenging of aerosol particles by snow crystals is defined as follows (Slinn, 1977; Pruppacher and Klett, 1978):

$$A(r_p) = \int_0^\infty K(r_p, r_c) N(r_c) dr_c \quad (4)$$

where $A(r_p)$ is the product of the collection kernel, $K(r_p, r_c)$, and the snow crystal size distribution, $N(r_c)$, integrated with respect to the equivalent snow crystal radius, r_c . Where r_c equals one half the snow crystal volume divided by its cross-sectional area. The collection kernel represents the number of aerosol particles of size r_p scavenged within the volume swept out by a falling snow crystal, r_c , for a given time period. The size distribution of each type of snow crystal, $N(r_c)$, is defined as the number of snow crystals of radius r_c to $r_c + dr_c$ per unit volume. Few expressions for the volume distributions of snow crystals

are currently available. The well-known Gunn and Marshall (1968) formula is:

$$N(r_c) = N_0 \exp(-\lambda r_c) \quad (5)$$

where $N_0 = 0.037P^{-0.87}$ (cm^{-3}) and $\lambda = 51P^{-0.48}$ (s^{-1}). Size distributions collected by Hobbs et al. (1972) indicate that for each snow crystal type (c) a distribution can be described by a gamma function such that the following applies:

$$N(r_c) = N_0 r_c^\alpha \exp(-\lambda r_c^\beta), \quad (6)$$

where N_0 is the initial crystal distribution and α , β , and λ are constants for each snow crystal type. The Gunn and Marshall distribution is a special case of the general gamma distribution, as is seen upon setting $\alpha = 0$ and $\beta = 1$.

The analytic expression for the collection kernel produced by snow crystals of arbitrary shapes is found directly from the solution of the particle convective diffusion equation in the presence of external forces (Wang, 1985). This flux formulation only holds for submicron particles; particle diffusivity breaks down for particles greater than one micron in radius. Particle flux density onto a falling snow crystal is expressed as:

$$\vec{j} = n(r_p) B \vec{F}_{\text{ext}} - \bar{f}_p D_p \nabla n(r_p) \quad (7)$$

where $n(r_p)$ is the size-dependent particle number concentration, whose flux is assumed to be conservative, $\nabla \cdot \vec{j} = 0$. Under steady-state conditions, with \vec{F}_{ext} nondivergent and conservative, eq. 7 leads to the particle convective-diffusion equation:

$$B \vec{F}_{\text{ext}} \cdot \nabla n(r_p) - \bar{f}_p D_p \nabla^2 n(r_p) = 0 \quad (8)$$

The net external force is:

$$\vec{F}_{\text{ext}} = \vec{F}_e + \bar{f}_h \vec{F}_{\text{th}} + \bar{f}_v \vec{F}_{\text{df}}$$

where \vec{F}_e , \vec{F}_{th} and \vec{F}_{df} are the electrostatic, thermophoretic, and diffusiophoretic forces, respectively. Forces due to turbulence, gravitation, and inertia are insignificant for this size range.

Particle mobility is defined as $B = C_{\text{sc}} / (6\pi\nu_a r_p)$, where ν_a is the dynamic viscosity of air and C_{sc} is the Cunningham slip correction factor:

$$C_{\text{sc}} = 1 + [1.26 + 0.4 \exp(-1.1r_p/\lambda_a)] \lambda_a / r_p$$

The mean free path, λ_a , is $6.6 \cdot 10^{-6} (T/273.15 \text{ K}) (1013.25 \text{ mb}/P)$, T and P are the ambient temperature and pressure. The Brownian diffusion coefficient is defined as $D_B = B k_b T$, where k_b is the Boltzmann constant. The particle, thermal, and vapor ventilation factors, \bar{f}_p , \bar{f}_h , \bar{f}_v , are calculated from the formulas proposed by Hall and Pruppacher (1977):

$$\begin{aligned}\bar{f}_v &= 1 + 0.14 x^2, & x < 1.0 \\ &= 0.96 + 0.28 x, & x > 1.0\end{aligned}\quad (9)$$

where $x = N_{Sc, \nu_a}^{1/3} N_{Re, r_c}^{1/2} \cdot N_{Sc, \nu_a}$ is the Schmidt number for water vapor, and N_{Re, r_c} is the Reynolds number for the characteristic dimension r_c . Hall and Pruppacher have indicated that under typical atmospheric conditions, $\bar{f}_h \sim \bar{f}_v$. The particle ventilation factor is computed as in eq. 9, except that the particle Schmidt number, N_{Sc, ν_p} , is used in place of the water vapor Schmidt number.

Aerosol particle charge is proportional to the square of the particle's radius and is multiplied by the charge strength q (Takahashi, 1973):

$$Q_p = q r_p^2 \quad (10)$$

Snow crystal charge is defined as the product of the reciprocal of capacitance and the gradient of surface potential, ϕ_e :

$$Q_c = V \phi_e / C_{ap} \quad (11)$$

Thus electrostatic forcing is described as:

$$\vec{F}_e = Q_p V \phi_e / C_{ap} \quad (12)$$

Thermophoretic forcing, \vec{F}_{th} , is dependent on the temperature gradient between the snow crystal surface and the ambient nonsaturated air. Brock (1962) has formulated \vec{F}_{th} as:

$$\vec{F}_{th} = C_{th} \nabla T \quad (13)$$

where:

$$C_{th} = -42/5\pi \nu_a r_p (k_{ap} + 2.5 N_{Kn}) / [(1 + 3 N_{Kn})(1 + 2k_{ap} + 5 N_{Kn})] D_T \quad (14)$$

The thermal conductivity of air, k_a , and the thermal conductivity of the aerosol particle, k_p , are expressed as a dimensionless ratio, $k_{ap} = k_a/k_p$. The thermal diffusivity coefficient is defined as $D_T = k_a/\rho_a c_p$, where c_p is the specific heat of air at constant temperature. The diffusiophoretic force, \vec{F}_{df} , is dependent on the water vapor gradient between the snow surface and the ambient nonsaturated air. Hidy and Brock (1970) expressed \vec{F}_{df} as:

$$\vec{F}_{df} = C_{df} \nabla \rho_v, \quad (15)$$

where:

$$C_{df} = -6\pi \nu_a r_p (1 + \sigma_{va} \chi_a) (D_{va} M_a) / (M_v \rho_a) \quad (16)$$

χ_a is the mole fraction of dry air, and σ_{va} is the surface tension for water in air; their product is approximately 0.26 (Schmidt and Waldmann, 1960). M_a is the molecular weight of dry air, and M_v is that of water. Water vapor diffusivity, D_{va} , is assumed constant at a given temperature and pressure. Values for $\rho_a(T)$ are from the Smithsonian Tables, while values for $k_a(T)$, $k_p(T)$, $\lambda(P, T)$,

$D_{va}(P,T)$, and $\eta_a(T)$ are from Pruppacher and Klett (1978). Under subsaturated conditions, \vec{F}_{th} is an inward (towards the ice crystal) directed force and \vec{F}_{df} is outward. Supersaturated conditions (ice crystal growth) are such that \vec{F}_{th} is an outward force and \vec{F}_{df} is inward. For submicron aerosols $|F_{th}| > |F_{df}|$ and therefore the net phoretic effect is an inward forcing for subsaturated conditions and an outward forcing for supersaturated conditions.

Conservation and nondivergence of the external forces:

$$\nabla \cdot \vec{F}_{ext} = Q_p/C_{ap} \nabla^2 \phi_e + C_{th} \nabla^2 T + C_{df} \nabla^2 \rho_v = 0$$

allow for the formation of a force potential, $\vec{F}_{ext} = -\nabla \phi$, that satisfies the Laplace equation:

$$\nabla^2 \phi = 0 \quad (17)$$

A solution to eq. 8 is obtained when eq. 17 and the required boundary conditions [$n(r_p) = 0$ at $\phi = \phi_0$, $n(r_p) = n(r_p)_\infty$ at ∞ , $\phi_\infty = 0$] are satisfied (Wang, 1985):

$$n(r_p) = n(r_p)_\infty \{ \exp[B(\phi - \phi_0)/D_p \bar{f}_p] / [\exp[B\phi_0/D_p \bar{f}_p] - 1] \} \quad (18)$$

The particle flux definition of the collection kernel:

$$[K(r_c, r_p, T, RH) = -1/n_\infty \delta / \delta t [\int_0^\infty n(r_p) dr_p]$$

is defined as:

$$K(r_c, r_p, T, RH) = B / [\exp(B\phi_0/D_p \bar{f}_p) - 1] \int_S \nabla \phi \cdot dS \quad (19)$$

The integral about an arbitrary snow crystal surface, S , can be evaluated by applying the Gauss theorem for an electrostatic analog to solve for the surface force potential, ϕ_0 (Houghton, 1949; McDonald, 1963; Pruppacher and Klett, 1978; Wang, 1985), as:

$$\begin{aligned} \int_S \nabla \phi \cdot dS &= -4\pi C_{ap} (\phi_0 - \phi_\infty) \\ &= -4\pi C_{ap} [Q_c Q_p / C_{ap} + \bar{f}_{th} C_{th} (T_s - T_\infty) + \bar{f}_{df} C_{df} (\rho_{v,s} - \rho_{v,\infty})] \end{aligned} \quad (20)$$

Substituting eq. 20 into eq. 19 gives the final form for the collection kernel:

$$K(r_p, r_c) = C_{ap}(r_c) \Phi(r_c, r_p, T, RH), \quad (21)$$

where:

$$\Phi(r_c, r_p, T, RH) = -4\pi B \phi_0 / [\exp(B\phi_0/D_p \bar{f}_p) - 1]$$

and is a function of r_c , r_p , T and RH . Relative humidity is defined here as the ratio of water vapor density to water vapor density at saturation ($RH = \rho_v / \rho_{v,sat}$).

The capacitance of snow crystals has been measured by Podzimek (c.f. Pruppacher and Klett, 1978) and McDonald (1963) for several ideal geometries. Capacitance is linearly proportional to r_c and can be described as:

$$C_{ap} = C_{cp} r_c \quad (22)$$

where C_{cp} (in esu units) is a surface area factor and is less than or equal to unity. For a thin disk $C_{cp} = 2/\pi$, for an oblate spheroid $C_{cp} = e/\sin^{-1}e$, and for a prolate spheroid $C_{cp} = e/\ln[z(1+e)]$, where z is the axis ratio (major axis/minor axis) and $e = (1 - 1/z^2)^{1/2}$. Podzimek has found that the capacitance of a hexagonal plate is within 4% of that of a thin disk. The capacitance of a solid prism (or a hollow prism) is within 12% (or 10%) of that of prolate spheroids. The capacitance of dendrites can be approximated from that of disks or oblate spheroids on the basis of the percentage of surface area to perimeter of a dendrite compared to that of a solid disk. Increasing the perimeter to describe finely detailed snow crystals allows charge density to appear in regions of high local surface curvature, thus increasing the crystal's ability to hold charge, water vapor, or temperature densities. Houghton (1949) suggested that capacitance be determined by the overall outline, implying that the fine structure has little effect. Fig. 1 indicates the relationship between capacitance and

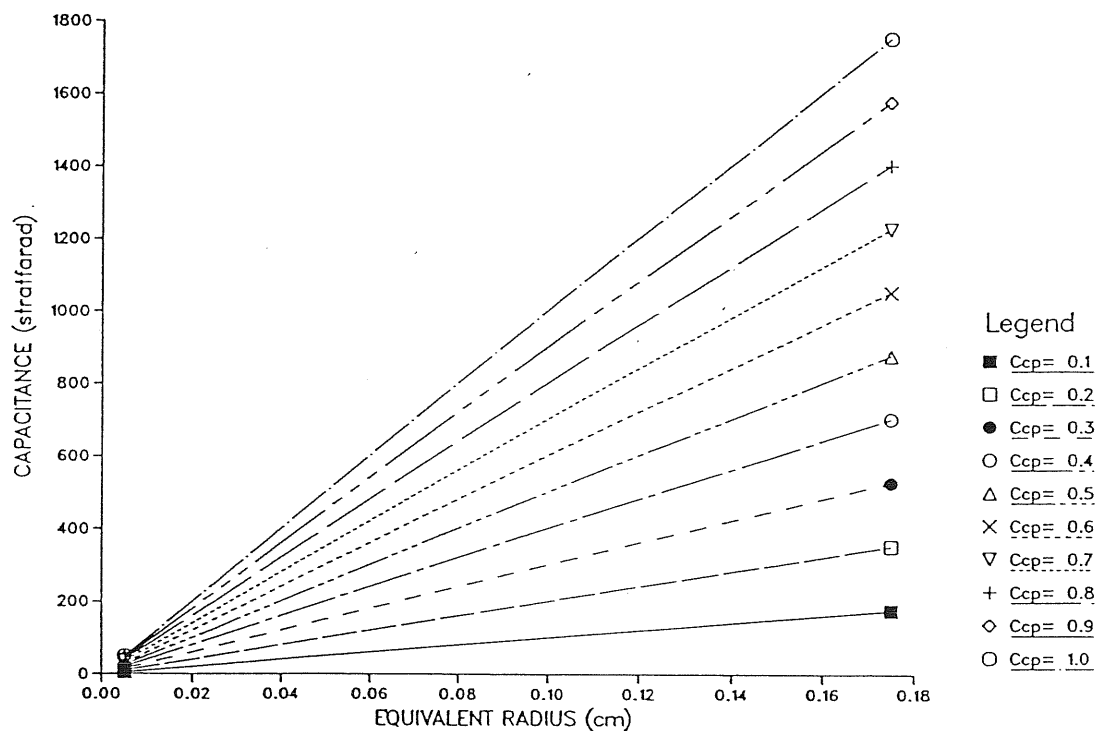


Fig. 1. Capacitance as a function of equivalent radius for snow crystals of various geometries as specified by C_{cp} .

equivalent radius for several surface area factors. The maximum factor ($C_{cp}=1$) is for a sphere; as the surface area-to-perimeter ratio decreases, so does C_{cp} .

The particle scavenging rate is solved by integrating eq. 4, using the gamma distribution given in eq. 6 and the collection kernel as defined in eq. 21:

$$A(r_p) = \int_0^\infty C_{ap}(r_c) \Phi(r_c, r_p, T, RH) N_0 r_c^\alpha \exp(-\lambda r_c^\beta) dr_c \quad (23)$$

For the case of uncharged snow crystals or aerosol particles (no electrostatic force), Φ is no longer a function of r_c and:

$$A(r_p) = \int_0^\infty \Phi(r_p, T, RH) C_{cp} r_c N_0 r_c^\alpha \exp(-\lambda r_c^\beta) dr_c \quad (24)$$

which upon integration has the form:

$$A(r_p) = N_0 C_{cp} \Phi(r_p, T, RH) \Gamma(\gamma) / (\beta \lambda^\gamma) \quad (25)$$

where $\gamma = (\alpha + 2)/\beta$, the gamma function is an infinite series of the form:

$$\Gamma(\gamma) = \lim_{n \rightarrow \infty} 1/\gamma \prod_{m=1}^n (1 + \gamma/m)^{-1} n^\gamma$$

Eq. 25 represents the general form for uncharged snow crystal scavenging for any crystal shape and gamma distribution. When electrostatic forcing is considered, this solution does not reduce to a gamma function but to a complicated infinite series, which at present has not been fully evaluated.

Applying the Gunn and Marshall distribution to eq. 25 results in a function of similar form to eq. 1:

$$\begin{aligned} A(r_p) &= N_0 C_{cp} \Phi(r_p, T, RH) / \lambda^2 \\ &= 122.3 P^{0.09} C_{cp} \Phi(r_p, T, RH) \end{aligned} \quad (26)$$

Eq. 26 appears to be an improvement over existing scavenging rate formulas, in that it describes scavenging rates as a function of relative humidity, temperature, particle size, and crystal shape. The total aerosol particle scavenging rate is the integral of eq. 23 with respect to particle size:

$$A = N_0 C_{cp} \Gamma(\gamma) / (\beta \lambda^\gamma) \int_0^\infty \Phi(r_p, T, RH) dr_p \quad (27)$$

DISCUSSION AND RESULTS

Particle scavenging rate variability due to snow crystal geometry is described in terms of the crystal capacitance. This has been linearized (eq. 22) as a function of equivalent radius, r_c , and the snow crystal surface factor, C_{cp} . Fig. 2 indicates scavenging rates for $0.01 \mu\text{m} < r_p < 1.0 \mu\text{m}$ with $0.1 \leq C_{cp} \leq 1.0$. Scav-

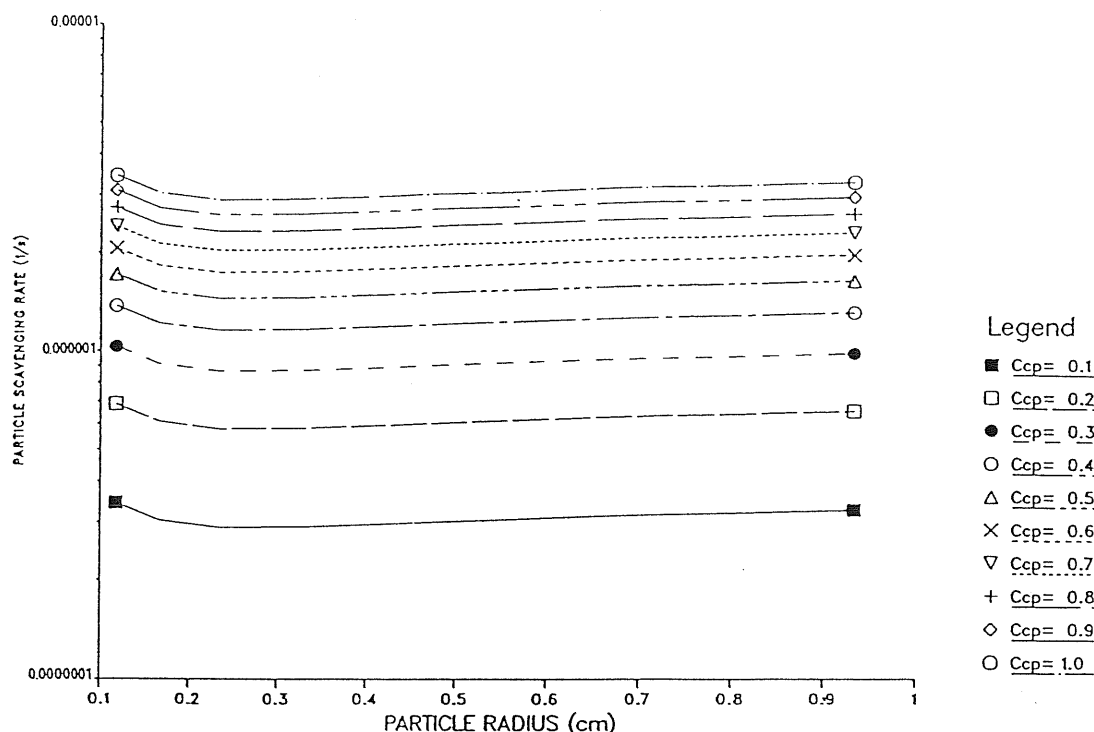


Fig. 2. Particle scavenging rate as a function of particle radius for snow crystals of various geometries as specified by C_{cp} at $T=263.15$ K, $RH=95\%$.

enging rates per snow crystal cross-sectional area are greatest for spherical shapes ($C_{cp}=1$), less for disk-like shapes ($C_{cp}=0.5$), and least for needles ($C_{cp}=0.1$). Values of C_{cp} for complicated crystal shapes can be estimated from simpler shapes or measured by making metal molds and measuring their capacitance (McDonald, 1963). The corresponding scavenging rates can then be calculated from eq. 25.

Increasing temperature increases the particle mobility and hence increases Brownian diffusion (Fig. 3). Temperature also increases C_{th} and thermophoresis, which is most significant for particles near $1.0 \mu\text{m}$ in size.

The effects of relative humidity are seen in Fig. 4. Thermodiffusiophoresis decreases rapidly as the environment approaches the saturated condition. At saturation, $\nabla T = \nabla p_v = 0$ and $\vec{F}_{th} = \vec{F}_{th} = 0$, leaving only Brownian diffusion as a scavenging mechanism in this size range (in the absence of electrical effects). The scavenging rate inflection occurs at about $0.25 \mu\text{m}$ at $0.93 < RH < 0.95$. For unsaturated conditions, the phoretic effect essentially fills in the Greenfield gap, the region where Brownian diffusion and inertia become insignificant. Certainly electrostatic effects would enhance this effect, as has been seen for aerosol particle-ice crystal scavenging efficiency studies (Wang and Prupacher, 1980; Martin et al., 1980; Wang, 1985; Miller, 1987; Miller and Wang, 1989).

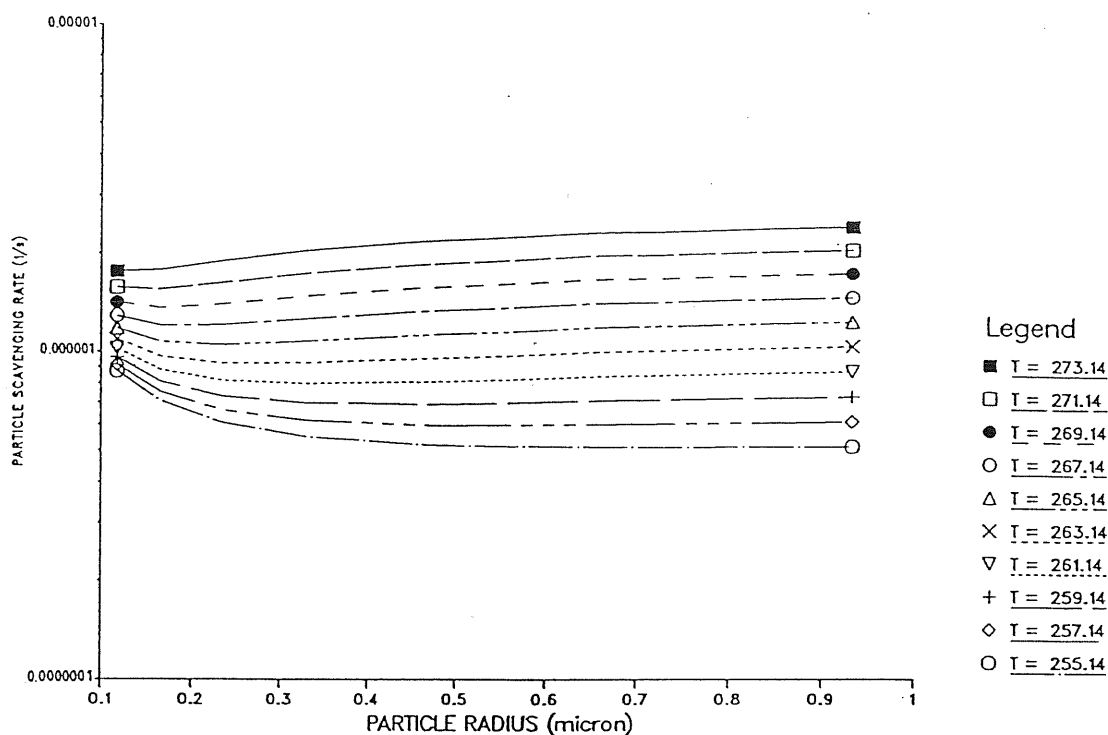


Fig. 3. Particle scavenging rate as a function of particle radius and temperature changes from at $RH=95\%$, $C_{cp}=1/\pi$.

Comparison of the present model (eq. 26) with the Knutson and Stockman model (eq. 1), the Lacaux and Warburton model (eq. 2), and the model of Slinn (eq. 3) indicates general agreement. The scavenging rate based on (eq. 2) with $D_p=0.03 \mu\text{m}$ ($A=1.26 \cdot 10^{-4}$) agrees well with the corresponding values from eq. 26: $A(r_p)=1.05 \cdot 10^{-4}$. Eqs. 1, 3 and 26 are compared in Fig. 5 for $D_p=0.5 \mu\text{m}$. The temperature is set to 270 K (25° F), and the relative humidity for eqs. 1, 2 and 3 is assumed to be near 100%. Eq. 26 is solved for two relative humidities, 95% and 99%, and for two geometries, spherical ($C_{cp}=1.0$) and disk-like ($C_{cp}=0.5$). Fig. 5 indicates that variations in relative humidity are much more significant than variations in crystal type. A 5% change in the relative humidity can cause a change of an order of magnitude in the scavenging rate, while a 50% change in capacitance will cause a change of a factor of two in scavenging rate. This suggests that for scavenging rates of aerosols in this size range, the relative humidity is the dominant mechanism over crystal shape when particles or crystals are uncharged.

The present model indicates a weak dependence on precipitation due to the choice of the Gunn and Marshall distribution where $\alpha=0$ and $\beta=1$ in the gamma distribution for snow crystals. Solutions for other Gaussian distributions may indicate significantly different precipitation dependence.

Data on snow crystal scavenging rates are very limited. Fig. 6 presents mea-

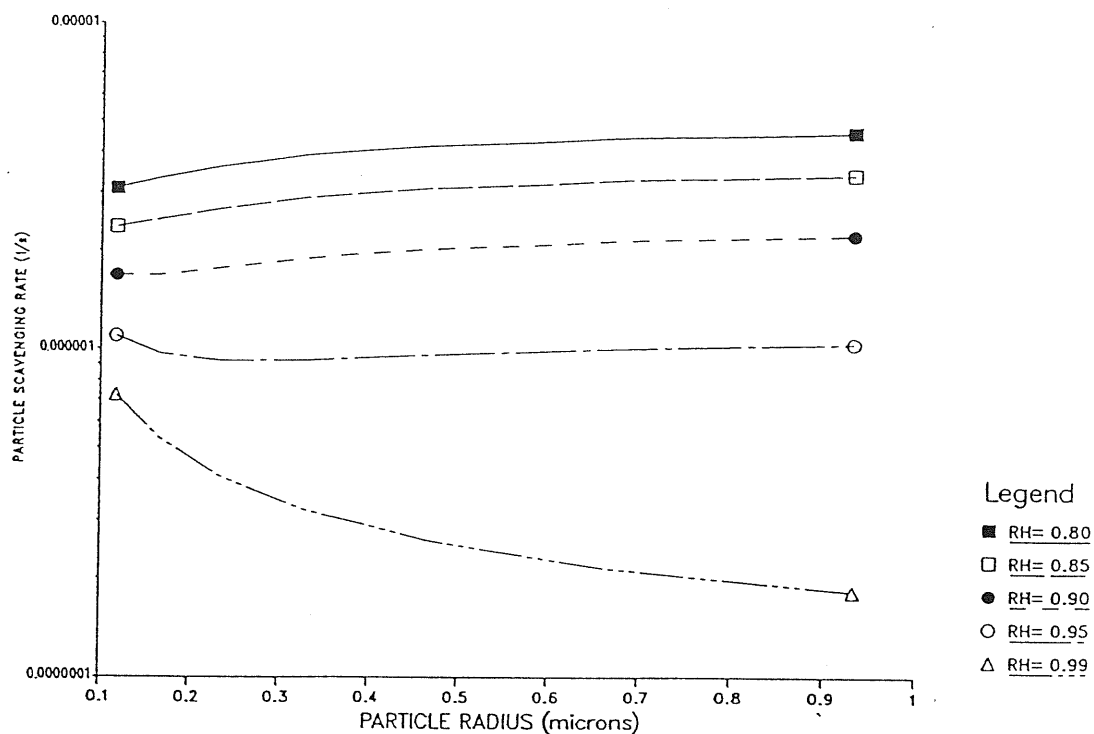


Fig. 4. Particle scavenging rate as a function of particle radius and relative humidity at $T=263.15$, $C_{cp}=1/\pi$.

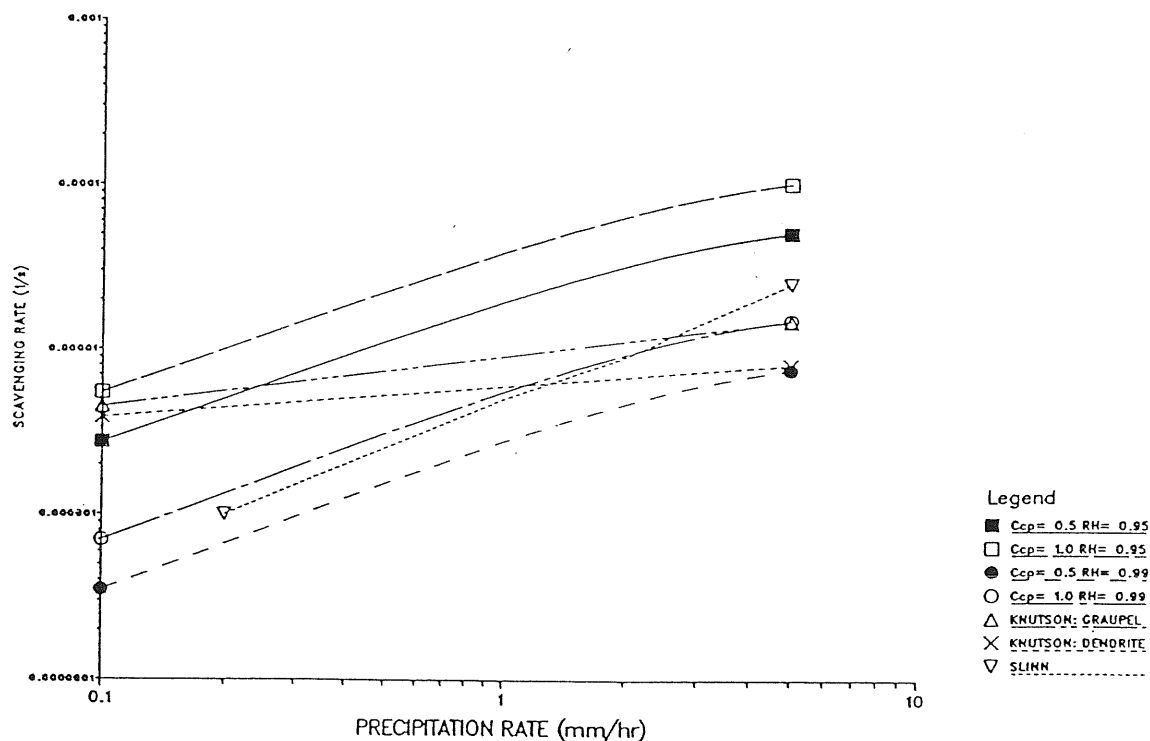


Fig. 5. Predicted scavenging rate of $0.5 \mu\text{m}$ particles as a function of precipitation rate from Knutson and Stockham (1974), Slinn (1984), and the present model.

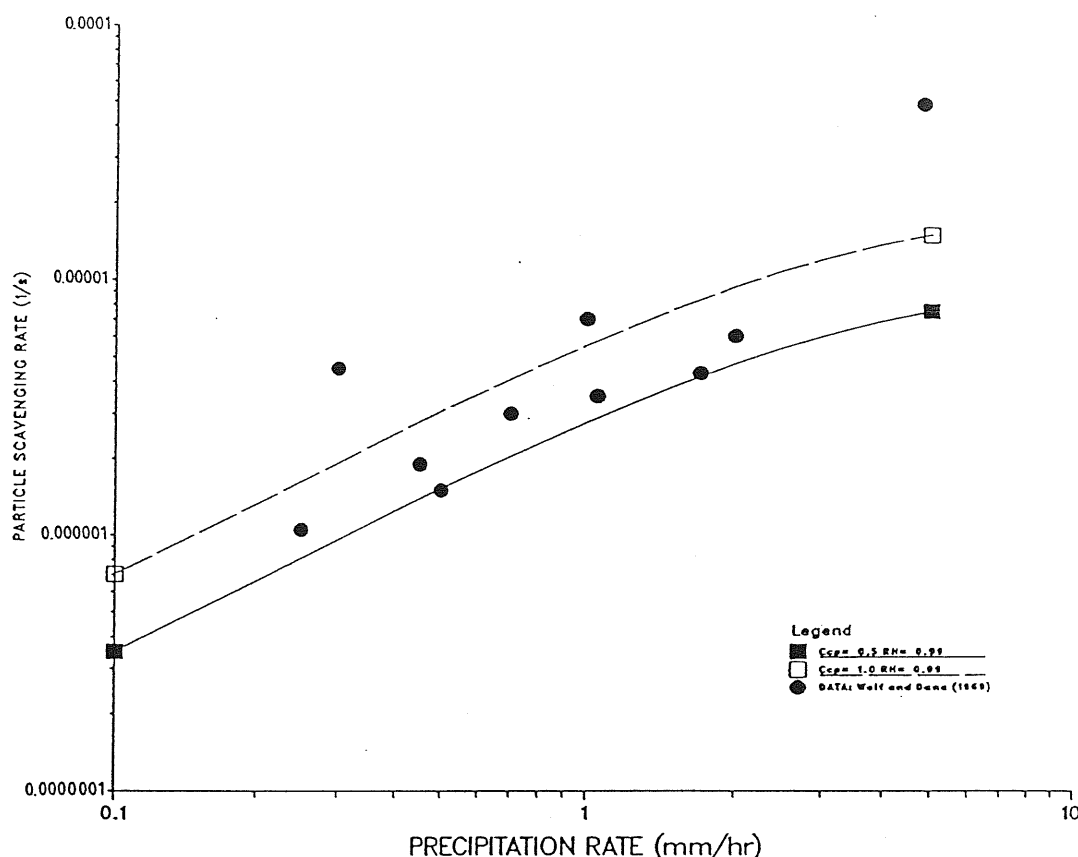


Fig. 6. Comparison of scavenging rate predicted by the present model with experimental results of Wolf and Dana (1969) for $0.5 \mu\text{m}$ particles as a function of precipitation rate.

sured data from Wolf and Dana (1969) $D_p = 0.5 \mu\text{m}$ at 269 K with relative humidity assumed to be near 100%. These data are consistent with the results of the present model. More studies both in the field and laboratory are necessary to validate this theory.

CONCLUSIONS

The solution of the particle convective-diffusion equation results in a collection kernel that is a function of the surface force potential. The surface force potential incorporates Brownian diffusion, thermodiffusiophoresis, and electrostatic charges. In the absence of electrostatic charge, the collection kernel is integrated with the most general form of the gamma distribution of snow crystals to give a generic solution of aerosol particle-snow crystal scavenging rates. Since this model does not consider inertia, $1 \mu\text{m}$ is the upper limit of particle size. This model extends existing scavenging rate models by including effects associated with relative humidity and capacitance.

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